Processing and Deposition of Nanocrystalline Oxide Composites for Thermal Barrier Coatings

Technical Report on ONR Grant No. N00014-95-1-0626
for the period of January 1, 2000-March 31, 2000

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1. Electrophoretic Deposition

As described in the previous report [1], electrophoretic deposition was used as the method in coating substrate materials for thermal barrier coating applications. In this report, we examined the effect of alumina on the thermal stability and electrical conductivity of 1.3 wt% yttria-stabilized zirconia (YSZ)-based thermal barrier coatings. In addition, the effects of powder calcination temperature and film thickness on the thermal stability of Al_2O_3-YSZ coatings were also examined.

In previous reports, we prepared Al_2O_3-YSZ nanocomposite powders using the colloidal mixing approach [1]. These powders were coated onto Ni substrates using electrophoretic deposition. The concentrations of the slurry components were the same as reported previously [1].

2. Thermal Stability of Alumina-Zirconia Thermal Barrier Coatings

2.1. Effect of Alumina Content

Coatings with various Al_2O_3 contents were prepared and tested for thermal stability (Table 1). Following electrophoretic deposition, the Al_2O_3-YSZ-coated Ni substrates were pretreated in argon at 1150 °C for 1 hr (ramp = 10 °C/min). The coated substrates were then tested for thermal stability by heating to 1150 °C in air (ramp =10 °C/min). The samples were examined visually every hour for visible spallation of the coating and oxidation of the Ni substrate. All experiments were repeated three times.

The addition of a small amount of Al_2O_3 to the coatings had a positive effect on the thermal stability. The coating without Al_2O_3 began to crack and spall after 3 hr of heat treatment. The most stable compositions were 5 wt% Al_2O_3-95 wt% YSZ and 10 wt% Al_2O_3-90 wt% YSZ, which lasted for 6 hr before significant spallation occurred. Coatings with higher Al_2O_3 contents (20 wt% and 100 wt%) had poor thermal stability, lasting only 1 hr at 1150 °C before spallation occurred. Figure 1 shows an optical micrograph of 5 wt% Al_2O_3-95 wt% YSZ before and after 6 hr of heat treatment.

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave Blank)*	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	31 Mar 2000	Progress Report: 1 Jan 00 - 31 Mar 00

4. TITLE AND SUBTITLE
Processing and Deposition of Nanocrystalline Oxide Composites for Thermal Barrier Coatings

5. FUNDING NUMBERS
G - N00014-95-1-0626

6. AUTHORS
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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Office of Naval Research
800 North Quincy Street
Ballston Tower One
Arlington, VA 22217-5660

10. SPONSORING / MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION / AVAILABILITY STATEMENT

Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

This report describes the thermal stability and electrical conductivity of nanocrystalline oxide composites for thermal barrier coating applications. Electrophoretic deposition was used to coat the nanocomposite powders onto nickel substrates. The effect of alumina content, powder calcination temperature, and film thickness on the thermal stability of zirconia-based coatings was examined. It was found that a small addition of alumina (5 wt%) and high calcination temperatures (1300 C) enhanced the thermal stability of the coatings. Additionally, 50 micron-thick coatings were more thermally stable than thinner coatings. The electrical conductivity of the powders was also measured to determine the effect of alumina on oxygen conductivity.

14. SUBJECT TERMS
Nanocrystaline Processing, Electrophoretic Deposition, Thermal Barrier Coatings

15. NUMBER OF PAGES
7

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

When more than 10 wt% Al_2O_3 was added to the YSZ-based materials, the thermal stability of the coatings would be reduced. This might be attributed to a difference in the thermal expansion coefficients of Al_2O_3 and YSZ, which could induce stress and subsequent failure of the coating. Thermal stabilization with the addition of 5-10 wt% Al_2O_3 could be explained by a decrease in oxygen conductivity (see section 3).

Coatings were examined with X-ray diffraction (XRD) to determine their crystal phase before and after the heat treatment. Figure 2 shows the XRD patterns of 5 wt% Al_2O_3-95 wt% YSZ before and after 6 hr of heat treatment at 1150 °C. Prior to heat treatment, ZrO_2 was present in the tetragonal phase. Ni peaks were noted from the Ni substrate. After 6 hr of heat treatment, ZrO_2 retained 100% tetragonal phase. NiO peaks were observed after the heat treatment, due to cracking of the coating and oxidation of the Ni substrate. Other Al_2O_3-YSZ nanocomposite coatings had similar phases present as this system before and after heat treatment.

2.2 Effect of Powder Calcination Temperature

5 wt% Al_2O_3-95 wt% YSZ powder was calcined at various temperatures for 8 hr prior to the coating process. Table 2 shows the thermal stability of the resulting coatings as a function of the powder calcination temperature. The most stable coatings were obtained with powders calcined at 1300 °C and 1400 °C, which lasted for 6 hr before significant spallation occurred at 1150 °C in air. Coatings produced with powders calcined at 1100 °C and 1200 °C lasted for 2 and 3 hr, respectively. Powders calcined below 1100 °C produced coatings that underwent immediate cracking upon pre-treatment at 1150 °C in argon, due to densification of the coated material.

2.3. Effect of Coating Thickness

The effect of film thickness on the thermal stability of Al_2O_3-YSZ coatings was investigated. A thick coating would provide the Ni-based substrate with additional thermal insulation but might suffer from poor mechanical stability. By increasing the deposition time, coatings with thicknesses of ~20 to ~50 μm were prepared and tested for thermal stability. The maximum coating thickness was ~50 μm with a deposition time of 240 s. Coatings thicker than ~50 μm proved to be mechanically unstable, as they cracked upon drying.

5 wt% Al_2O_3-95 wt% YSZ powders were calcined at 1300 °C prior to deposition. The thermal stability of the resulting coating increased slightly as the coating thickness increased. ~50 μm-thick coating sustained 6 hr at 1150 °C in air. Coatings of 30 μm and 40 μm thick were stable for 4 hr, while a 20 μm-thick coating was stable for only 3 hr.

3. Electrical Conductivity of Alumina-Zirconia Composites

One of the primary failure modes for thermal barrier coatings was severe oxidation of the bond coat, which would result in the spallation of the thermal barrier

coating. This oxidation was caused by oxygen diffusion through the zirconia-based coating. At temperatures > 500 °C, oxygen diffusion through zirconia became relatively fast through ionic conduction [2]. Oxygen adsorbed on the surface of zirconia in the form of oxygen ions was rapidly transported through the zirconia structure. By lowering the oxygen conductivity of YSZ coating, the oxidation rate of the underlying bond coat could be potentially decreased and thermal stability of the coatings would be improved. Secondary components, such as alumina, could be added to YSZ to decrease its oxygen conductivity and diffusivity. A small amount of alumina additive might greatly decrease the ionic conductivity of YSZ, without significantly increasing the thermal conductivity of the thermal barrier coating.

Previous researchers have found that the ionic conductivity of Al_2O_3-YSZ depended heavily on the processing conditions and the starting powders used [3-10]. Drennan and Badwal [5] reported that YSZ containing 20 wt% Al_2O_3 had higher total ionic conductivity than pure YSZ. Other researchers have shown a decrease in ionic conductivity upon addition of Al_2O_3 to ZrO_2 [7].

In this study, the electrical conductivity of the Al_2O_3-YSZ composites was measured to determine the role of alumina in oxygen ionic conductivity and to help explain the results from the thermal stability studies. For the conductivity studies, Al_2O_3-YSZ powders were compacted to pellets (1 cm diameter x 0.5 cm thickness) with 30,000 psi at 25°C, and were calcined at 1400 °C for 8 hr. Platinum electrodes were painted onto the pellet surfaces, and calcined at 800 °C for 1 hr. DC conductivity of the resulting pellets was measured using the two-point probe method with a Keithley 236 power supply over the temperature range of 300 °C to 900 °C.

Figure 3 shows resistivity (ohms·m) as a function of temperature for several compositions. The resistivity increased as the alumina content was increased. At 900 °C, pure YSZ had a resistivity of 4.0 kohms·m, while 10 wt% Al_2O_3-90 wt% YSZ had a resistivity of 57.0 kohms·m. The conductivity results were consistent with the findings of Mori *et al.* [6]. Alumina acted as an insulator in the Al_2O_3-YSZ composite, decreasing the oxygen conductivity. The results of the conductivity experiments verified that the addition of a small amount of alumina (≤ 10 wt%) had a positive effect on the thermal stability of the coatings by decreasing oxygen conductivity without sacrificing mechanical stability.

4. Future Work

For future studies, a NiCrAlY bond coat will be applied onto the Ni substrate via electrophoretic deposition prior to depositing the 5 wt% Al_2O_3-95 wt% YSZ coatings. The thermal stability of the NiCrAlY/Al_2O_3-YSZ layered coating will be investigated. The thickness and pretreatment of the NiCrAlY bond coat will be optimized to maximize the thermal and mechanical stability of the thermal barrier coatings. The NiCrAlY/Al_2O_3-YSZ coatings will be heat treated at elevated temperatures (1000 °C–1400 °C) for extended periods, and will be subjected to thermal cycles of 25 °C-1150 °C-25 °C (with ramping and cooling rates of ~300 °C/min) to examine any failure modes.

5. References

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Table 1. Coating lifetime for various alumina-zirconia nanocomposites. The coating lifetime was given by the duration before spallation was first observed for a coating heat treated at 1150 °C in air.

Coating Composition	Coating Lifetime (hr)
100 wt% YSZ	3
5 wt% Al_2O_3-95 wt% YSZ	6
10 wt%Al_2O_3-90 wt%YSZ	6
20 wt% Al_2O_3-80 wt% YSZ	1
100 wt% Al_2O_3	1

Table 2. Effect of powder calcination temperature on the thermal stability of the resulting ~50 μm-thick 5 wt% Al_2O_3-95 wt% YSZ coatings.

Calcination Temperature (°C)	Coating Lifetime (hr)
1100	2
1200	3
1300	6
1400	6

Table 3. Effect of coating thickness on the thermal stability of 5 wt% Al_2O_3-95 wt% YSZ at 1150 °C in air.

Coating Thickness (microns)	Coating Lifetime (hr)
20	3
30	4
40	4
50	6

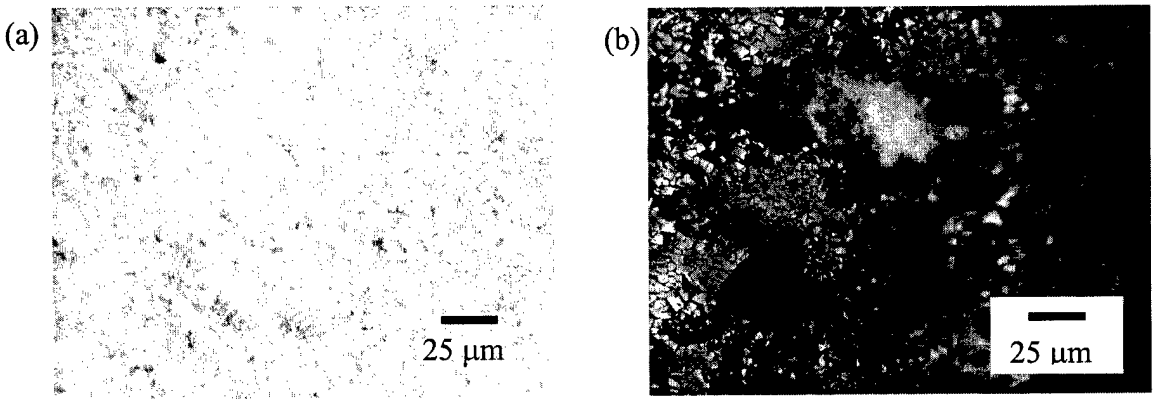

Figure 1. 5 wt% Al_2O_3-95 wt% YSZ (a) before and (b) after 6 hr of heat treatment at 1150 °C in air. Cracks are noted in the coating after the heat treatment.

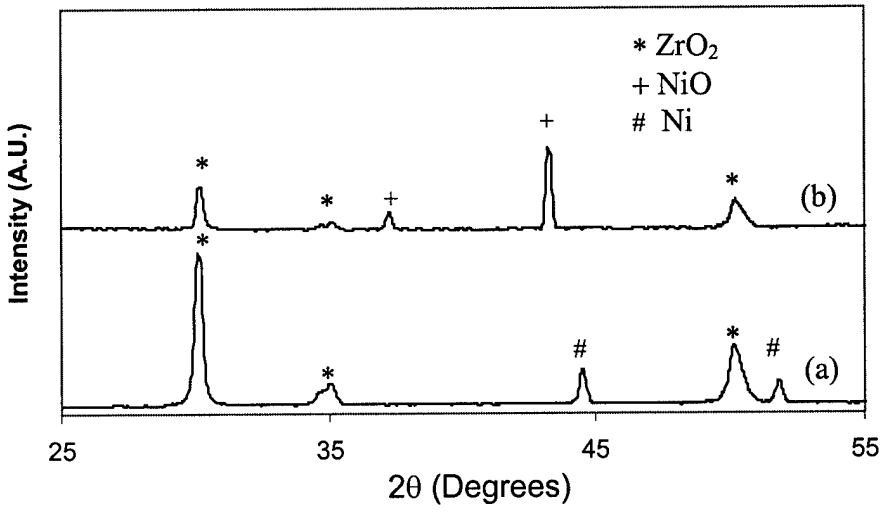

Figure 2. X-ray diffraction patterns of 5 wt% Al_2O_3-95 wt% YSZ (a) before and (b) after 6 hr of heat treatment at 1150 °C in air.

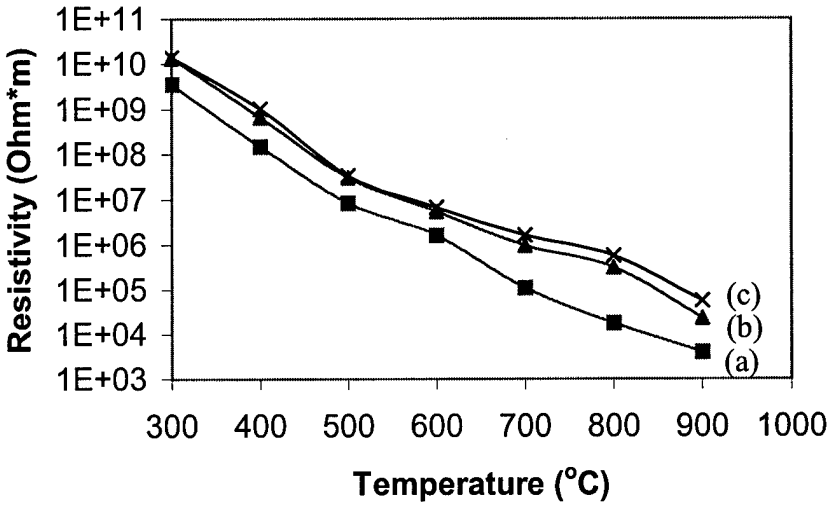

Figure 3. Resistivity as a function of temperature for coatings of (a) YSZ, (b) 5 wt% Al_2O_3-95 wt% YSZ, and (c) 10 wt% Al_2O_3-90 wt% YSZ.